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RESPONSE UNDER 37 CFR 1.116
EXPEDITED PROCEDURE

IN THE U.S. PATENT AND TRADEMARK OFFICE

February 26, 2003

Applicants: Kazuo NAKAMURA et al

For: BORON-DOPED ISOTOPIC DIAMOND AND PROCESS FOR PRODUCING THE SAME

Serial No.: 09/732 799 Group: 1765

Confirmation No.: 2965

Filed: December 8, 2000 Examiner: Kunemund

Atty. Docket No.: OPS Case 421A

Assistant Commissioner for Patents
Washington, DC 20231

APPELLANTS' BRIEF ON APPEAL

Sir:

This is an appeal from the decision of the Examiner dated September 23, 2002, finally rejecting Claims 17-32.

REAL PARTY IN INTEREST

Tokyo Gas Co., Ltd. and Tokyo Gas Chemical Co., Ltd. are the assignees of the present application and the real parties in interest.

RELATED APPEALS AND INTERFERENCES

There are no related appeals or interferences to the present application.

STATUS OF CLAIMS

Claims 17-19 and 21-32 are pending in the present application and are the claims under consideration on appeal. Claims 1-16 and 20 have been canceled.

STATUS OF AMENDMENTS

An Amendment After Final Rejection incorporating the subject matter of Claim 20 into Claim 18 and canceling Claim 20 is being filed concurrently with this appeal brief. Additionally, the Amendment After Final Rejection changes the

dependency of Claim 27 to Claim 18 to address the Examiner's rejection of Claim 27 under 35 USC 112.

SUMMARY OF THE INVENTION

Appellants' invention, as defined by independent Claim 17, is directed to a single crystal diamond p-type semiconductor having a thermal conductivity of from about 26-31 W/cm°K and consisting of at least 99.5% isotopically pure ^{12}C or ^{13}C and boron in an amount not exceeding 100 ppm (specification page 5, lines 7-17).

Appellants' invention, as defined by independent Claim 18, is directed to a method of manufacturing a single crystal diamond p-type semiconductor having a thermal conductivity of from about 26-31 W/cm°K and a boron content not exceeding 100 parts per million (specification page 5, lines 7-25). The inventive method comprises the steps of providing a carbonaceous material containing isotopically purified ^{12}C or ^{13}C, providing a flux containing a nitrogen getter, adding boron into the carbonaceous material or/and the flux, or around the carbonaceous material and the flux, and diffusing the carbonaceous material into the flux under a high temperature and pressure to form a boron-doped single crystal diamond p-type semiconductor on a seed crystal diamond (originally presented Claim 8).

Claim 19 limits Claim 18 in requiring that the isotopically purified ^{12}C or ^{13}C have a purity of at least 99.5% (specification page 5, lines 7-9).

Claim 21 limits Claim 18 in requiring that the carbonaceous material is at least one member selected from the group consisting of pyrolytic carbon, a diamond synthesized by chemical deposition and carbon synthesized by chemical deposition (specification page 8, lines 25-28).

Appellants' invention, as defined by independent Claim 22, is directed to a single crystal diamond having a boron content and consisting of boron in an amount not

exceeding 100 ppm and at least 99.5% isotopically pure ^{12}C or ^{13}C (specification page 5, lines 7-13).

Appellants' invention, as defined by independent Claim 23, is directed to a process for producing a single crystal diamond containing boron comprising the steps of providing a flaky pyrolytic carbonaceous material, providing a metal solvent containing a nitrogen getter, adding boron to the flaky pyrolytic carbonaceous material or/and the metal solvent, dissolving the flaky pyrolytic carbonaceous material in the metal solvent and precipitating a single crystal diamond doped with boron in an amount not exceeding 100 parts per million on a seed crystal diamond (specification page 9, lines 22 and 23, originally presented Claim 8 and specification page 5, lines 18-21).

Claim 24 limits Claim 23 in requiring that the flaky pyrolytic carbonaceous material have a content of isotopically pure ^{12}C or ^{13}C or at least 99.5% (specification page 6, lines 22-24).

Claim 25 limits Claim 23 in requiring that the single crystal diamond doped with boron is precipitated at a side of the metal solvent having a lower temperature than a side of the solvent having a higher temperature (specification page 7, lines 10-20).

Claim 26 limits Claim 23 in requiring that the single crystal diamond doped with boron precipitates from the metal solvent under conditions of a high temperature and pressure (specification page 4, lines 13-21).

Claim 27 limits Claim 18 in requiring that the carbonaceous material be formed by pressing pyrolytic carbon powder in a steel die, introducing the pyrolytic carbon powder into a graphite capsule and heating the pyrolytic carbon powder in an induction heating furnace under a vacuum and at a temperature of 1,800-2,000°C to anneal the pyrolytic carbon powder (specification page 9, lines 11-18).

Claim 28 limits Claim 23 in requiring that the diamond is doped with boron in an amount of up to 60 ppm (specification page 5, lines 18-22).

Claim 29 limits Claim 23 in requiring that the diamond is doped with boron in an amount of up to 30 ppm (specification page 5, lines 18-25).

Claim 30 limits Claim 23 in requiring that the single crystal diamond doped with boron is a p-type semiconductor (specification page 5, lines 10-13).

Claim 31 limits Claim 23 in requiring that the single crystal diamond doped with boron have a thermal conductivity of from about 26-31 W/cm°K (specification page 5, lines 13-15).

Claim 32 limits Claim 22 in requiring that the diamond have a thermal conductivity of from about 26-31 W/cm°K (specification page 5, lines 13-15).

ISSUES

The first issue presented for review is whether Claim 27 is unpatentable under 35 USC 112, first paragraph. The second issue for review is whether Claims 17-22 and 32 are unpatentable under 35 USC 103(a) over Tsuji et al in view of Anthony. The third issue presented for review is whether Claims 23-31 are unpatentable under 35 USC 103(a) as being unpatentable over Tsuji et al in view of Anthony and Nakamura et al.

GROUPING OF CLAIMS

The claims do not all stand or fall together. Claims 17, 22 and 32 are directed to one invention, Claims 18, 19, 21, 23-26 and 28-31 are directed to a second separately patentable invention and Claim 27 is directed to a third separately patentable invention.

ARGUMENT

Claim 27 has been rejected by the Examiner under 35 USC 112, first paragraph, as not being supported by the original specification. The amendment accompanying the appeal brief addresses this rejection by changing the dependency of Claim 27 to Claim 18.

The presently claimed invention is based on the discovery that an isotopic diamond can be doped with boron and still possess a high thermoconductivity as well as be a p-type semiconductor. It is conventionally thought that boron atoms impede phonon conduction far greater than ^{13}C atoms in a ^{12}C diamond crystal or ^{12}C atoms in a ^{13}C diamond crystal. The main impedement to thermoconductivity of a diamond is phonon scattering caused by the mass difference, 1.00 atomic weight, between ^{13}C atoms and ^{12}C atoms. On the other hand, the average atomic weight of boron is 10.81 and it is different by 1.19 atomic weight from ^{12}C and by 2.19 atomic weight to ^{13}C. Moreover, boron atoms work as acceptors in the diamond crystal and 0.1% thereof is positively charged at room temperature. Due to this, the conventional thought was that atoms of boron would impede phonon conduction far greater than ^{13}C atoms in a ^{12}C diamond crystal or ^{12}C atoms in ^{13}C diamond crystals.

In contrast to the conventional thinking, the present inventors discovered that the thermoconductivity of isotopically purified diamond is hardly lowered by containing boron if the purity of the ^{12}C or ^{13}C diamond crystal is not less than 99.5% and the boron concentration is not more than 100 parts per million. A concentration of boron of no more than 100 parts per million is sufficient to obtain a p-type conductivity at room temperature, and yet enables the diamond to be used in electronic parts as a heat sink material.

The p-type single crystal diamond semiconductor of the present invention can be prepared by a high temperature/high pressure method in which a flux of molten metal is used to dissolve a carbonaceous source for producing the semicrystal

diamond. The molten metal takes up the carbon source up to its saturation concentration before a single crystal diamond precipitates therefrom. An advantage of the present invention is that an inexpensive flaky pyrolytic carbonaceous material can be used which is much less expensive than prior art carbonaceous sources and is soluble in the molten metal at a higher temperature than diamond so the diamond can precipitate out of the molten metal solution separately from the pyrolytic carbon. It is respectfully submitted that the prior art cited by the Examiner does not disclose the presently claimed invention.

The Tsuji et al reference is directed to a method of synthesizing single diamond crystals using a carbon source containing at least 99.9 atomic percent carbon-12. This process requires the graphitizing of carbon-12 to form a highly crystalline material which can be used as a carbon source in an ultra-high pressure creating apparatus to produce single diamond crystals through a temperature difference process. As stated by the Examiner, this reference does not disclose the doping of the isotopically pure diamond produced there with boron or that a p-type semiconductor can be produced thereby.

Column 4, lines 59-67 of this reference discloses that impurities lower the thermoconductivity of diamonds and that although the effect of nitrogen is small, improvements in thermoconductivity can be obtained if the diamond impurity is completely eliminated. Therefore, nothing in this reference suggests that any advantage would be gained by the addition of boron thereto and, in fact, this reference teaches away from the presently claimed invention in that it states that impurities are undesirable in the single diamond crystals produced there. It would be expected that the thermoconductivity of the isotopic single crystal diamond would be impaired due to the doping compound interfering with the crystalline structure.

The Anthony et al reference is directed to a stress-relieved chemical vapor deposition diamond produced by annealing the chemical vapor deposition diamond at a temperature above about 1,100 to about 2,200°C in a non-oxidizing atmosphere at a low pressure or vacuum and for a suitable short period of time which decreases with increasing annealing temperature so as to prevent graphitization of the diamond.

Anthony et al discloses in Column 6, lines 43-55, that additives may be contained in the starting CVD diamond films produced by deposition on substrates. Moreover, nitrogen, boron, oxygen and phosphorus may be present in the CVD diamond films in the form of intentional additives. Boron is disclosed as an intentional additive that can be used to reduce the intrinsic stress in CVD diamond films or to improve the oxidation resistance of the film. This reference goes on to state that lower levels of impurities tend to favor desirable properties of toughness and wear resistance and the most preferred films contain less than 5 parts per million and preferably less than 1 part per million impurities and intentional additives. Additionally, the boron concentration range of from 1 to 4,000 parts per million includes the concentration region exceeding 100 parts per million in which an isotopic effect would not occur according to the present invention. Therefore, this reference has no consideration regarding the improvement of thermoconductivity caused by doping isotopically purified diamond with boron in the claimed range to obtain a p-type semiconductor.

In Anthony et al, the boron is added to reduce intrinsic stress in a CVD diamond film or to improve the oxidation resistance thereof. The single diamond crystals of Tsuji et al are produced by a temperature difference process and not by chemical vapor deposition as required by Anthony et al. Therefore, it is respectfully submitted that one of ordinary skill in the art would not be motivated to add boron to the

single diamond crystals of Tsuji et al in that they are not produced by chemical vapor deposition and that it is expressly disclosed there that impurities are to be avoided. For this reason, it is respectfully submitted that article Claims 17, 22 and 32 are patentably distinguishable over the combination of Tsuji et al and Anthony et al.

The Nakamura et al reference has been cited by the Examiner in combination with the previously discussed references to reject Claims 23-31 in that Nakamura et al has been cited as teaching the use of flaky pyrolytic carbon as a source of diamonds. However, as stated above, one of ordinary skill in the art would not be motivated to add boron, as disclosed in Anthony et al, to the process of Tsuji et al in that Tsuji et al does not use chemical vapor deposition to form the single diamond crystals disclosed there. Particularly since the boron is added in Anthony et al to solve a problem caused by the process of forming the diamond by chemical vapor deposition while Tsuji et al forms the diamonds by a completely different process. As such, Claims 18, 19, 21, 23-26 and 28-31 are separately patentable in that they are directed to processes for producing a single crystal diamond p-type semiconductor containing boron which is not suggested by the combination of Tsuji et al, Anthony et al and Nakamura et al.

Claim 27 is felt to be even further patentably distinguishable over the cited references in that it requires additional processing steps be performed on the pyrolytic carbon powder before it is used to make the single crystal diamond p-type semiconductor of the present invention. Since these additional processing steps are not suggested in any of the references cited by the Examiner, it is respectfully submitted that Claim 27 is even further patentably distinguishable thereover.

Although the Examiner has not made a showing of prima facie obviousness under 35 USC 103, Appellants respectfully

submit that objective evidence is contained in the present specification which clearly establishes the patentability of the presently claimed invention over the prior art cited by the Examiner. In the Examples and Comparative Examples contained on pages 12-21 of the present specification, natural diamonds, natural diamonds which are boron doped, and isotopic diamonds are compared with diamonds according to the present invention, which are isotopic diamonds doped with boron. As can be seen by the results contained in Tables 2-5 of the present specification, the diamond single crystals according to the present invention had an advantageous combination of properties of a high thermoconductivity and also were p-type semiconductors. This is clearly unexpected in light of the prior art cited by the Examiner and further establishes the patentability of the presently claimed invention thereover.

CONCLUSION

For the reasons advanced above, it is respectfully submitted that the patentability of the presently claimed invention has been established. Reversal of the Examiner is respectfully solicited.

Respectfully submitted,

IN TRIPLICATE

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Encl: Appendix
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APPENDIX

17. A single crystal diamond p-type semiconductor having a thermal conductivity of from about 26-31 W/cm°K and consisting of at least 99.5% isotopically pure ^{12}C or ^{13}C and boron in an amount not exceeding 100 ppm.

18. A method of manufacturing a single crystal diamond p-type semiconductor having a thermal conductivity of from about 26-31 W/cm°K and a boron content not exceeding 100 ppm comprising the steps of:

providing a carbonaceous material containing isotopically purified ^{12}C or ^{13}C;

providing a flux containing a nitrogen getter;

adding boron into the carbonaceous material or/and the flux, or around the carbonaceous material and the flux; and

diffusing the carbonaceous material into the flux under a high temperature and pressure to form a boron-doped single crystal diamond p-type semiconductor on a seed crystal diamond.

19. The method of Claim 18, wherein the isotopically purified ^{12}C or ^{13}C has a purity of at least 99.5%.

21. The method of Claim 18, wherein said carbonaceous material is at least one member selected from the group consisting of pyrolytic carbon, a diamond synthesized by chemical deposition and carbon synthesized by chemical decomposition.

22. A single crystal diamond having a boron content and consisting of boron in an amount not exceeding 100 ppm and at least 99.5% isotopically pure ^{12}C or ^{13}C.

23. A process for producing a single crystal diamond containing boron comprising the steps of:

providing a flaky pyrolytic carbonaceous material;

providing a metal solvent containing a nitrogen getter;

adding boron to the flaky pyrolytic carbonaceous material or/and the metal solvent;

dissolving the flaky pyrolytic carbonaceous material in the metal solvent and precipitating a single crystal diamond doped with boron in an amount not exceeding 100 ppm, on a seed crystal diamond.

24. The process of Claim 23, wherein the flaky pyrolytic carbonaceous material has a content of isotopically pure ^{12}C or ^{13}C of at least 99.5%.

25. The process of Claim 23, wherein the single crystal diamond doped with boron is precipitated at a side of the metal solvent having a lower temperature than a side of the solvent having a higher temperature.

26. The process of Claim 23, wherein the single crystal diamond doped with boron precipitates from the metal solvent under conditions of a high temperature and pressure.

27. The process of Claim 18, wherein the carbonaceous material is formed by pressing pyrolytic carbon powder in a steel die; introducing the pyrolytic carbon powder into a graphite capsule and heating the pyrolytic carbon powder in an induction heating furnace under a vacuum and at a temperature of 1,800 to 2,000°C to anneal the pyrolytic carbon powder.

28. The process of Claim 23, wherein the diamond is doped with boron in an amount of up to 60 ppm.

29. The process of Claim 23, wherein the diamond is doped with boron in an amount of up to 30 ppm.

30. The process of Claim 23, wherein the single crystal diamond doped with boron is a p-type semiconductor.

31. The process of Claim 23, wherein the single crystal diamond doped with boron has a thermal conductivity of from about 26-31 W/cm°K.

32. The single crystal diamond of Claim 22, wherein said diamond has a thermal conductivity of from about 26-31 W/cm°K.